

TRANSMITTAL FORM FOR FILING PATENT APPLICATION

Attorney

Docket No.: VER-114XX

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BOX PATENT APPLICATION

Assistant Commissioner for Patents

Washington, D.C. 20231

Date: April 29, 1999

First Named Inventor or
Application Identifier: Marc-Olivier Coppens

Sir:

Transmitted herewith under 37 CFR § 1.53 for filing is the patent application of:

Inventor: Marc-Olivier Coppens

Entitled: METHOD FOR OPERATING A CHEMICAL AND/OR PHYSICAL PROCESS BY MEANS OF A
HIERARCHICAL FLUID INJECTION SYSTEM

[] This is a request for filing a [] **continuation** [] **divisional** [] **continuation**
in-part application under §1.53(b) of prior Application No. _____, filed
 _____ entitled:

Enclosed are:

[X] 25 pages of written description, claims and Abstract, inclusive

[X] 3 sheets of [X] informal [] formal drawings of Figs. 1-3 (one set)

[X] Oath or Declaration

[] Newly executed (original or copy)

[] Copy from prior application (37 CFR 1.63(d)) (for continuation/divisional).

The entire disclosure of the prior application, from which a copy of the oath
 or declaration is supplied, is considered as being part of the disclosure of
 the accompanying application and is hereby incorporated by reference therein.

[X] To be filed later ✓

[] Cover sheet and Assignment of the invention to:

[] Certified copy of a _____ application (if foreign priority is
claimed) with letter claiming priority under Rule 55.

[] Information Disclosure Statement with ___ citations

[] Preliminary amendment is enclosed.

[X] Return receipt postcard

[] Other:

[] Verified statement was filed in prior application. Status still proper and
desired

JC526 U.S. PRO
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04/29/99

jc660 U.S. PRO
 04/29/99

TRANSMITTAL FORM FOR FILING PATENT APPLICATION (CONTINUED)

Attorney

Docket No.: VER-114XX

- ☐ Priority is claimed under 35 USC § 120 as indicated on the attached sheet 4.
- ☐ Priority is claimed under 35 USC §119(a)-(d) as indicated on the attached sheet 4.
- ☐ Priority is claimed under 35 USC §119 (e) as indicated on the attached sheet 4.
- ☐ _____ is hereby appointed Associate Attorney by:
Registration No.:

Attorney of Record

Registration No.:

- ☐ **Power of Attorney** in the originally-filed application has been granted to one or more of the registered attorneys listed below. The attorneys listed below not previously granted power in the originally-filed application, as well as _____, are hereby given associate power:

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- ☐ Cancel in this application original claims _____ of the prior application before calculating the filing fee. (At least one original independent claim must be retained for filing purposes.)

- ☐ Add in this application claims _____ per amendment before calculating fee.

CLAIMS FILED:	MINUS BASE:	EXTRA CLAIMS:	RATE:	BASIC FEE:
				\$760.00
Independent	4 - 3	= 1	x \$78.00 =	78.00
Total	13 - 20	= 0	x \$18.00 =	0
<input type="checkbox"/> Multiple Dependent Claims (1st presentation)			+ \$260.00 =	0
SUBTOTAL FILING FEE				\$838.00
Small Entity filing, divide by 2. (Note: verified statement must be attached per \$1.9, \$1.27, \$1.28.)				0
TOTAL Filing Fee				\$838.00

- ☐ The filing fee has been calculated above; a check in the amount of _____ is enclosed.

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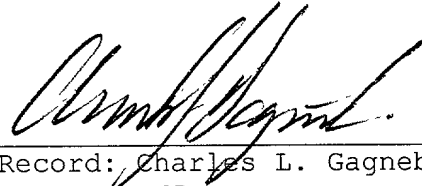
Attorney Docket No.: VER-114XX

TRANSMITTAL FOR FILING PATENT APPLICATION (CONTINUED)

- [X] In the event a Petition for Extension of Time under 37 CFR §1.17 is required by this paper and not otherwise provided, such Petition is hereby made and authorization is provided herewith to charge Deposit Account No. 23-0804 for the cost of such extension.
- [] The Commissioner is hereby authorized to charge payment of any additional filing fees under 37 CFR §1.16 associated with this communication or credit any overpayment to Deposit Account No. 23-0804.

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TRANSMITTAL FOR FILING PATENT APPLICATION (CONTINUED)

☐ Priority is claimed under 35 USC § 120 of prior Application(s)
No. _____, filed _____, entitled:

☐ The above-identified application(s) is/are assigned of record to:

☐ Priority is claimed under 35 USC § 119 (a)-(d) of the following application(s).

(Application Number) (Country) (Filing Date)

(Application Number) (Country) (Filing Date)

(Application Number) (Country) (Filing Date)

☐ The above-identified application(s) is/are assigned of record to:

☐ Priority is claimed under 35 USC § 119 (e) of the following provisional application(s).

(Application Number) (Filing Date)

(Application Number) (Filing Date)

(Application Number) (Filing Date)

☐ The above-identified provisional application(s) is/are assigned of record to:

☐ The claim of small entity status in the above-identified provisional application(s) is made in this application and a copy of the small entity form(s) from the provisional application(s) is/are enclosed.

SUBMIT IN TRIPLICATE

CLG:mes/203808

Title: Method for operating a chemical and/or physical process by means of a hierarchical fluid injection system

5 The invention relates to a method for operating a chemical and/or physical process, e.g. a fluid bed reaction, and especially deals with the injection of fluids, such as gases, liquids and/or suspensions in a controlled typically uniform way. More in detail, the chemical and/or physical processes are carried out within a vessel containing a fluid or fluidized medium, which medium can be a gas and/or a liquid, optionally in combination with solid particles. The fluids are injected using an injection device consisting of
10 tubes or channels that are connected in a hierarchical fashion so that the fluid entering a first channel is divided into channels of the same or different diameter and length, each or some of which are further divided into channels of the same or different diameter and length, and so on. The
15 injection system can also comprise combinations of such tree-like or fractal-like elements, embedded in a plane (two-dimensional) or in space (three-dimensional).

Multiphase reactors or multiphase contacting devices, are commonly applied in process industry. In such processes
20 two or more fluids have to be brought into contact with each other to efficiently transfer one or more components from one phase into another phase, where the reaction or contacting process occurs. This is in particular so for transport limited processes, of which there are many in industrial
25 practice.

The invention can be used for gas/solid fluidized bed, gas/liquid and other processes. A typical application is the reduction and uniformization or otherwise control of bubble sizes to optimize the operation of fluidized bed,
30 slurry and gas/liquid reactors. The embodiments of the present invention will depend upon the nature of the fluids and the application.

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An important example is a fluidized bed reactor, where gas is injected through a distributor plate in the bottom of a vessel to fluidize a bed of solid particles that are to react with the gas or catalyze a chemical reaction
 5 between gas molecules. Gas bubbles grow from the bottom to the top of the bed as a result of the pressure difference over the bed, simply because of the thermodynamics. The movement of the fluidized solid particles and the bubbles is turbulent, chaotic, and strongly dependent on the operating
 10 conditions.

Fluidized bed reactors are of considerable economic importance. An example of a large scale process that is preferably carried out as a fluidized bed process is the Fischer-Tropsch synthesis of liquid fuel from synthesis gas,
 15 which can be obtained by steam-oxygen gasification of coal or other hydrocarbons. This process involves the contacting of the gas stream with solid catalyst particles to produce the fuel:



Another important example in which similar problems occur is a gas/liquid process carried out in a bubble column or stirred tank reactor. The gas moves up from a distributor
 25 through a reactor vessel. The movement of the bubbles is again extremely complex. Typically, the shape and size of the bubbles cannot be or is hardly controlled and bubbles coalesce and grow as a result of the pressure difference between the top and the bottom of the reactor. A mixing
 30 device and fixed internals may be installed inside the vessel to improve the mixing by increasing the turbulence in the bed. Similar complex hydrodynamics exist for other processes in which several fluids or fluidized media are contacted to react or catalyze reactions.

35 To optimize the reaction process, the interfacial area between the fluid phases should be maximized or otherwise controlled. With the presently used reactor systems

or physical contacting devices, this forms a serious problem, especially because the hydrodynamics, as described above, are so complex.

Mixers can be used to increase turbulence and contacting, but they consume costly energy. In other cases, such as gas/solid fluidized beds, mixers may not be used. In addition, mechanical problems arise when mixing devices are employed.

Homogenization, i.e., achieving equal conditions at every point in the reactor vessel, is difficult to achieve. Imposing particular concentration or partial pressure profiles is even more difficult or for most reaction systems impossible. The existence of dead volumes, where no or much less reaction occurs, because of decreased local flow, is often inevitable.

Another problem in current realizations of such multiphase reactors and other processes involving several fluids or fluidized media that need to be contacted is 'scaling up' of these devices. A process is usually first investigated on a smaller scale, e.g. on lab scale, bench-scale or pilot scale, and then needs to be scaled up to the typically much larger industrial scale. Rules to scale up multiphase processes are typically empirical or at least semi-empirical and the errors are very large, because the processes are influenced by the hydrodynamics. With the present methods, it is a serious challenge to maintain similar hydrodynamics during scale up from the small to the large scale. In most cases, this is even impossible.

Whenever in this specification and the appended claims reference is made to a 'vessel' it is to be understood that this refers to a container for fluids and optionally particles, in which optionally provisions are made to feed additional fluids and/or particles, and optionally provisions are made to remove fluids and/or particles. Preferably such a vessel is operated in a continuous process. The examples of processes mentioned above, are all carried out in vessels.

In U.S. Patent 4,537,217 a fluid distributor is disclosed having a distribution surface with a plurality of uniformly spaced distribution openings, which is particularly suitable for application in chromatography.

10 U.S. Patent 5,354,460 discloses a fluid transfer system for obtaining uniform liquid distribution in industrial scale fluid transfer systems which accommodate plug flow operations.

25 Finally, Kearney (in: Fractals in Engineering, INRIA Proceedings, June 25-27, 1997, Arcachon, France) describes a three-dimensional mixing unit, which consists of a recursive structure of pipes. This structure is suitable for emulating turbulence using laminar flows.

35 It has now been found that the above mentioned
problems can be solved by introducing a fluid in said vessels

by means of a hierarchical network of channels comprising parent and child generations of channel formations, wherein substantially each channel in a parent generation is divided into N channels of the child generation, which network
5 terminates in channel exits, such that said fluid is discharged from the channel exits substantially uniformly throughout the vessel volume.

By discharging the introduced fluid substantially uniformly throughout the vessel volume, as is enabled by the
10 network structure, it was found that the operation of the processes which were carried out in said vessels is improved considerably. The introduction of fluids through said network results in equal pressures at all outlets of the tree. When the fluid is a gas, this leads to equal or otherwise
15 controlled bubble sizes, distributed as directed by the position of the outlets inside the vessel. A substantial uniform discharge of the fluid is intended to refer to a situation in which the discharge conditions with respect to local pressure and/or fluid velocity at substantially all
20 channel exits is substantially the same.

The network of channels according to the invention is a recursive structure, in which different generations of channel formations can be distinguished. When reference is made to a 'parent generation', a formation of channels having
25 a certain geometry and a certain size is meant. Each formation can comprise one or more channels or groups of channels, which channels are essentially of the same size or which groups of channels comprise channels of the same size in essentially the same orientation, and the geometry of
30 which channels or groups of channels are the same and these channels or groups of channels are arranged in any mutual order within each formation, although usually the arrangement within each formation will be regular or otherwise well defined. A 'child generation' is intended to be a formation
35 of channels, which channels are substantially similar in geometry to the channels of the corresponding parent

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generation from which it is branching, and either of the same or smaller size.

Substantially each channel of a parent generation branches into N channels which are members of its child generation. The value of N will depend on the process and what is practically achievable. According to the present invention, N is at least 2. The upper limit of N will be strongly determined by what is practically achievable, typically N will be smaller than about 8, preferably smaller than 6.

One of the surprising features of the present invention is that when N is not the same for all channels, for example when one of the channels is clogged, resulting in stopped flow of all the channels belonging to its child generation or generations, this does not necessarily adversely influence the process. Therefore, according to the invention, some variation in the value of N throughout the network is acceptable, depending on the application and the generation of channels it refers to, as long as substantially each channel branches into N channel. This means that according to the present invention a small fraction of the channels may branch in less than N channels, or even into no channels at all, without departing from the general concept of the present invention.

The injection devices which are used in the present invention comprise successive generations of channels, the first generation of channels being fed by a formation which is not necessarily a hierarchic structure. The last generation (bearing the highest number) comprises the channels that discharge in the vessel.

The number of generations will depend on the type of process. For a given application the person skilled in the art will be able to establish the required number of generations to construct a network that fills the volume of the vessel in an adequate manner. Usually, for most practical

applications the number of generations does not exceed about 10.

Channels are defined here as structures capable of transporting fluids, such as pipes, hoses or any duct.

5 In a preferred embodiment of a method according to the present invention, the network is a self-affine network of channels, i.e. a network wherein for several generations, the ratio of successive lengths is constant for channels of different generations running in the same direction. The
10 ratio of successive diameters in a self-affine network is also constant, but the length and diameter ratios need not be the same. In this preferred embodiment each of the channels in the parent generation have a diameter d_j and length l_j and each of the channels in the child generation have a diameter
15 d_{j+1} and length l_{j+1} , wherein the ratios d_j/d_{j+1} and/or the ratios l_j/l_{j+1} are substantially constant for channels of successive generations running in parallel direction. In this embodiment said network can also be a self-similar network which is a type of said self-affine network wherein the
20 ratios d_j/d_{j+1} and l_j/l_{j+1} are substantially constant for channels of successive generations independent of their direction.

By employing a self-affine network, optimal use can be made of the available volume of the vessel, which often
25 has dimensions that are not equal in all directions. A self-similar network would be especially useful in processes which are carried out in vessels which have more or less equal dimensions in all directions.

The networks of channels in the method of the present
30 invention can also be combined, viz. it is possible to construct networks comprising sub-networks that are self-affine or self-similar without departing from the general concept of the present invention. Such a combined network is therefore not necessarily self-affine or self-similar, but as
35 long as it comprises elements that are at least substantially

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self-affine or self-similar, the effects of the present invention can be obtained.

Although self-affine and self-similar structures are preferred embodiments of the present invention, and although
5 the requirement of self-affinity or self-similarity results in a structure wherein all exits have similar dimensions, . deviations from this definition can exist in networks according to the invention, without departing from the general concept of the present invention. Such 'non-perfect'
10 structures can still provide the desired effect described herein. In fact it is sometimes desirable to introduce such deviations from the strict self-affine and self-similar structures on purpose, for example in order to further minimize the influence of gradients, such as pressure and/or
15 concentration gradients, which often inevitably exist in vessels in which the networks according to the present invention are applied. By carefully varying the length and diameter of the channels of the network, in particular the diameter of the channels near the exits, gradients in factors
20 such as local bubble size, pressure drop and/or hydraulic path length can be imposed and consequently an even better control of the process is possible.

This feature which enables enhanced controlling and becomes possible with the hierarchical fluid injection system
25 according to the present invention will be referred to as to 'otherwise control' the processes wherein it is applied. It was found that the variations in exit diameters could be introduced quite simply by plugging the exits of the network with different plugs having holes of varying diameter.

30 Strictly speaking, self-affine or self-similar embodiments of the present invention require that each successive generation comprises channels that are in the same direction as the corresponding channels in the parent generation, or that the child channels are regularly rotated
35 with respect to their corresponding parent channels, the rotation being the same for each successive generation. It is

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to be understood that small deviations from this strict requirement need not influence the applicability of the channel network. In general, the influence of such deviations from this strict self-affinity or self-similarity with respect to orientation, becomes less in higher generations, i.e. in generations closer to the exits. This can be convenient in practical applications, since it allows flexibility in applying the network injection devices according to the present invention.

It has also been found that especially suitable networks for application in a method according to the present invention can be obtained when such networks are constructed such that the ratios of diameters and/or lengths of channels in successive generations of said network are related to N by

$$N = (d_j/d_{j+1})^A, \text{ and/or}$$

$$N = (l_j/l_{j+1})^D,$$

wherein A and D represent an integer or a real positive number.

It has been found that the control of flow or diffusion through the channels is especially effective when the above mentioned relations are fulfilled. The method according to the invention can be easily optimized by using these relations, as will be explained hereafter.

Preferably the value of D is between 2 and 3. In this case the resulting network will make use of the volume it extends in, while at the same time avoiding interference with the fluid inside the vessel which may be in motion and leaving enough space for the multiphase process that takes place inside the vessel.

Preferred processes in which any of the above mentioned networks can be successfully applied, are fluidized bed processes, slurry processes, gas/liquid bubble column processes and aeration processes, since all of these processes suffer from the problems set out in the above and

can therefore benefit from the method of the present invention.

For instance aeration processes are carried out in an aerator which contains an aqueous liquid phase in which a solid phase biomass is present. The biomass must be kept in suspension and a certain degree of turbulence must be maintained. Oxygen is supplied by bubbling air through the reactor. By applying a network as set out in the above, optimal mixing can be obtained, even without employing moving parts, and at the same time the formation of too large air bubbles is avoided.

Another surprising advantage of the present invention is the ease of scale up of processes. A method for scaling up according to the present invention comprises building a small scale vessel in which a fluid is introduced through a hierarchical network of channels which can be any network as described in the above, such that said fluid is discharged from the channel exits substantially uniformly throughout the vessel volume and determining optimal geometry and optimal values for the parameters N , Δ and D , and subsequently building a large scale vessel in which said geometry and parameters are substantially the same as in the small scale vessel. According to this method N , D and Δ are kept constant but the number of generations as well as the absolute respective lengths and/or diameters may differ to suit the demand of the large scale application.

The network according to the invention could be modified to further control the processes wherein it is applied, by having at least one material present near the exits of said network and/or said material is present as a coating on at least part of the inner surface of said network, which material is capable of chemical and/or physical interactions with fluids. Such materials could e.g. be a catalyst, an ion exchanger and/or an adsorbent. Examples are zeolites or ion exchange resins.

As mentioned in the above, further control can also be obtained by providing to any of the above mentioned networks means for obtaining a gradient in the dimensions of the respective exits. It was found that the variations in exit diameters could be introduced quite simply by plugging the exits of the network with different plugs having holes of varying diameter. When applied in a bubble column for instance, such an injection system could provide a means to further control the uniformity of the process by imposing the bubble sizes. When applied in fluidized beds, a gradient in the exit diameters of the channels of the hierarchic channel network system could provide a means to further control the uniformity of the process, since bubbles introduced near the bottom could reach diameters that, in the absence of any intrinsic pressure gradient resulting from the fluid column, would be substantially larger than the bubbles that are introduced near the top of the column.

The method according to the present invention enables optimized production using the above mentioned processes in combination with hierarchic network channels.

The networks of the present invention can be constructed easily by a variety of methods. For example, T- or Y-shaped elements can be connected easily by all kinds of means, such as gluing, soldering, welding, clamping, etc. Such networks can be easily extended by simply adding additional elements.

Suitable materials for constructing the networks according to the present invention are all materials that are essentially inert to the reaction conditions, e.g. stainless steel, or other metal, glass, quartz glass, etc.

It is also possible to use a bundle of flexible channels, which are subsequently formed to the desired shape, viz. having their exits at the desired location in space. In such a network of all similar channels, $\Delta = 2$.

It should be mentioned that the actual structure of the hierarchical network is quite different for different

processes. A channel may split into N channels, of which the lengths and diameters are the same or, usually, smaller than that of the parent channel. Each of these channels splits in the same or different way. A typical, yet not the only possible, division is such that, within a certain range of scales, a self-similar or self-affine network results: for several generations, the ratio of successive lengths is constant. The ratio of successive diameters is also constant, but the length and diameter ratios need not be the same. The ratios of diameters will control the flow or diffusion through the channels. Assume, e.g., a division of each channel of diameter d_j into N new channels of diameters d_{j+1} . The ratio d_{j+1}/d_j is typically maintained constant over several generations. Then, the diameter exponent Δ can be defined as follows:

$$(d_j)^\Delta = N(d_{j+1})^\Delta \text{ or } N = (d_j/d_{j+1})^\Delta$$

If the diameter exponent $\Delta=2$, the flow rates are the same through successive constructions, because the flow cross-section is the same throughout: when all the small branches of a certain generation are taken together then the sum of cross-sectional areas of all channels belonging to a certain generation is the same as the cross-sectional area of the preceding generation.

When Δ is 3, however, a complete volume is reached, but the flow is slowed down through the network. Δ may take intermediate values with corresponding effect. If all channel diameters are the same, Δ becomes infinitely large.

Optimal values of Δ depend on the type of process. A person skilled in the art will be able to determine, optionally aided by routine experiments optimal values for each process.

The set of outlet points of all the channels is, in the limit for an infinite number of generations, a fractal, with a fractal dimension D defined by the self-similar

branching. If a finite subset, and possibly all points, are reached within the (embedding) space in which the network lies, then the fractal dimension D equals the embedding dimension, i.e., 3 for a volume and 2 for a structure within a plane. For a 'network' consisting of a single channel, D would become zero because there is only one outlet. By controlling the branching ratio N and the ratio of successive lengths of the elements or channels that are combined in the network, l_{j+1}/l_j , the fractal dimension D , defined by:

$$(l_j)^D = N(l_{j+1})^D \text{ or } N = (l_j/l_{j+1})^D$$

may be modified to take fractional values in between zero and the embedding dimension (2 or 3). In this way, through D and Δ and the relative orientation of successive units in the hierarchy, a variety of structures can be formed.

Advantages such as robustness, trivial scalability and/or ease of construction of such hierarchical designs for technical applications as described above have not been perceived until now. The above cited prior art relates to the uniform distribution or collection of a liquid by means of a planar (two-dimensional) device in which all the channels have the same lengths or form a hierarchical network of a well-defined, specific structure disclosed in the patent. Each point in the plane would be reached, were the constructions described in the patent continued ad infinitum, hence $D=2$. At the outlets, the liquid pours out or is collected in a uniform way; typical applications are in chromatography.

The prior art structures are not suitable or optimal for the present applications to which the present invention relates, for which other D and Δ may and will usually be better, and combinations of hierarchical structures can be employed. Also, the devices described in the present patent are used inside a three-dimensional or possibly quasi-two-

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dimensional vessel (i.e., a vessel where one of the dimensions is much less than the other two), and not on top or below it. Applications are typically in the field of multiphase reactor engineering, although not limited to the latter. If $D=2$, every point in the plane will be reached, yet the complete available surface area is filled. This may be optimal for a distributor, yet not for an injector inside a vessel, where a large free volume needs to remain available for the reactions, absorption or other physico-chemical processes.

Another device according to the present invention can be used to distribute a fluid in a controlled and typically uniform way, so that the pressure drop from the inlet or inlets to the multitude of outlets is the same or otherwise controlled through the geometry of the structure. This device has the following design. A first channel, through which a fluid enters the device, splits in N_1 channels of the same or lower diameter and length, each of which again splits in N_2 channels of the same or lower diameter and length, and so on, until a smallest scale. Several such hierarchical constructions may be linked into a larger spatial structure: in this case, one main inlet pipe typically branches out into N_0 hierarchical fluid injection structures. The complete injection device is placed inside a vessel; depending on its size, if necessary, it is attached to the walls of the vessel.

Although the disclosed family of structures to which the claims relate is more general, (fractal) self-similar and self-affine designs are particularly attractive because of the ease of scaling up such structures. A self-similar structure is one where parts of the structure are rescaled copies of the whole; this is in principle only possible for true fractals with an infinite scaling range - the self-similarity is therefore interpreted to be valid only down to a smallest scale, the inner cutoff. For a self-affine structure, the similarity ratios of elements in different

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directions is different; they appear like stretched self-similar structures. Self-affine structures are also scaleable and may be attractive alternatives to self-similar structures for vessels that have different dimensions in different
 5 directions (e.g., a cylinder with a diameter different from its height). Easy scalability and facilitated construction in a wide range of sizes are two results of this hierarchical structure. It suffices to add additional generations in the hierarchical cascade(s) to make a larger structure, without
 10 changing the flow pattern. These are some of the features that make the networks of channels according to the present design attractive.

The largest and smallest sizes of the channels in the hierarchical fluid injection system depend on the particular
 15 application and on the size of the vessel. The fluid outlets are usually at the last stage. They may have a special process-dependent shape or structure that is different from all other channels and contain a porous plug, non-catalytic, or catalytic material, depending on the application.

20 The branching ratios of the hierarchical fluid injection system, N_1 , N_2 , etc. need in principle not be equal to each other, but in a self-similar or self-affine scaleable structure (the preferred embodiment for many applications), they are all equal and denoted by N .

25 Another interesting possibility is that $N^I = N_1 = N_2 = N_3 = \dots$ and $N^{II} = N_4 = N_5 = N_6 = \dots$, so that $N = N^I \times N^{II} =$ constant. Obvious generalizations hereof are also possible.

Two or more successive branchings may actually be considered as belonging to the same (double) generation,
 30 which makes the final design also self-similar or self-affine, apart from the inlet channel or 'trunk' of the tree.

By the definition given above, the ratio of the sizes of successive elements is constant and the same in all directions for a self-similar design, while in a self-affine
 35 design, the generation independent ratio is different in different directions (e.g., the vertical and the horizontal

directions). Again, the trunk, connecting different trees, is the natural exception. The diameter exponent Δ and the fractal (similarity) dimension D , defined earlier, are two characteristic numbers for the hierarchical construction, as they are directly related to the ratio of successive channel diameters and lengths respectively.

Their values are very important to achieve a uniform or otherwise controlled injection of fluid into the vessel containing the second fluid. They are strongly dependent on the application. In general, high values of Δ are suitable for bulkier structures that fill more space, but the resistance to flow through the structure is low. High values of D may also typically yield bulkier structures, although the bulkiness may be compensated by a lower value of Δ . When D is higher, the inlets reach a larger fraction of the vessel, yet less space is available for the fluid inside the vessel, which may not be desirable. The optimum of D , Δ , the number of generations, and the branching ratios, is clearly determined by the problem at hand, viz. the process to be operated, yet the global geometrical properties of the hierarchical structure claimed through this patent are similar and described as above; all these structures belong to the same family.

The facile control over the distribution of the fluid injection point locations and of the constant or otherwise controlled injection flow rates of the injected fluid, means that a physico-chemical process inside the vessel can be geometrically controlled in a unique way. Homogenization or uniformization of flow is usually attempted by mixing, which involves mechanical devices that consume energy and are more complex to build and maintain than the present invention. Although a hierarchical fluid injector may be used as a fluid injecting mixer, it does in its typical operation mode not involve moving parts, presenting another advantage over conventional designs. Another advantage is that a hierarchical unit can also be very robust against clogging,

which is especially important when solids are involved in the process.

Brief description of the drawings

5 It is to be understood that the shown embodiments only serve as good examples to carry out the invention in particular situations, yet that the structures to which this application relates are much more general, as described above.

10 Figure 1 shows a two-dimensional projection of a quasi-two-dimensional fluid injection device that can be used to inject secondary gas in a gas/solid fluidized bed. When the device is put inside a bed of solid particles, fluidized at minimum fluidization by a distributor under the bed,
15 fluidization is uniform and the bubbles are of a constant and controlled size, under a wide range of operating conditions.

The shaded areas indicate channels belonging to the first generation (1) and channels belonging to the third generation (3).

20 Figure 2 shows a two-dimensional projection (looking top down) of a three-dimensional fluid injection device that can be used to inject secondary gas in a gas/solid fluidized bed or in a gas/liquid reactor.

25 Figure 3 shows the same fluid injection device as in Figure 2 but under a different angle.

The invention will now be illustrated by the following examples, which are added for illustration only and are not intended to limit the scope of the invention in any
30 sense.

Example 1

A device as depicted in Figure 1 was assembled out of Plexiglas tubes to observe and illustrate the capabilities of
35 a hierarchical fluid injection system. It consisted of a central tube, which divided into 2 tubes that further divided

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into 2 tubes, all in the same plane so as to form an H-shape connected to the central tube. The horizontal bar of the H was as long as each of the vertical bars. Four hollow H-shapes, twice as small as the just described H, were
 5 connected to the open extremities of the latter H. All tubes in this example had the same diameter, were hollow and contained a hole in the middle to connect to a tube of the previous generation. The connections were made using glue and such that there were no leaks. The entire network measured
 10 about 30 cm.

The hierarchical construction was submerged in a fluidized bed of solid particles. The bed was fluidized at minimum fluidization (the particles were just fluidized as an emulsion phase) using an air stream through a distributor
 15 below the bed, i.e., the common way of operating a fluidized bed. Secondary air was injected through the hierarchical fluid injection system. Under a wide range of conditions (tested until 6 times the minimum fluidization rate), the bed was fluidized with small uniformly distributed bubbles. Air
 20 distribution through the lower distributor only, gave rise to a swarm of very large and smaller bubbles, which coalesced. Additional air injection through the described injection device changed the movement from highly chaotic to nicely controlled and uniform, as desired. Even after partial
 25 clogging of some tubes by solid particles, the unit functioned in an excellent way, demonstrating its robustness.

Example 2

A three-dimensional hierarchical fluid injection
 30 device according to Figures 2 and 3 was assembled from tubes of the same materials as in the previous example, yet they did not have the same size. A vertical central tube divides into 6 tubes, branching out horizontally with their endpoints on a regular hexagon. From the endpoint of each tube descends
 35 another tube. In the present example, each of the latter tubes branched into 3 tubes in the same plane. This

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'generating unit' repeated through a self-similar cascade. From the endpoints of each of these tubes, said generating unit was connected vertically up and vertically down. The vertical tubes were connected. The 3 horizontal tubes of the upper generating unit were oriented in a direction directly opposed to that of the tubes of the lower unit, and the tubes are parallel to the tubes forming the hexagon mentioned above. This procedure was continued in a recursive way. Fluid could flow from the inlet through the structure to and through each of the extremities of the smallest tubes. The diameter of the network was 35 cm.

Diameter and length ratios of the tubes are variable and depend on the particular application. For a gas/liquid process, it was found that a low value of the diameter exponent Δ is favorable, while for a gas/solid process a higher value with a few generations is best. In the shown self-similar design, by means of example, the fractal dimension was $D = \log 6 / \log 2 \approx 2.58$. In the described implementation, a projection of the setup will give a hexagonal grid, as shown in the figure: the dimension of this projection is 2, which means that bubbles of a gas flowing through the structure may rise through the vessel and reach a maximum volume, even while the total structure does not occupy too much space, as indicated by the low value of the fractal dimension, substantially lower than 3.

Example 3:

A structure similar to Figure 1, about 10 cm in size, was built out of hollow metal channels with a diameter down to 100 μm and submerged horizontally in a vessel filled with water. The diameter exponent Δ of the structure was 2 and the structure consisted of 3 generations. It was observed that when air was blown through the horizontal structure, bubbles of the same size rose from the 64 exits. Moreover, even at relatively high gas rates, the bubbles rose in nice parallel streets without visible coalescence either horizontally or

vertically. Such uniform behavior is highly desirable for gas/liquid absorption, aeration and other reactor processes, because it makes the mass transfer of components between the gas and liquid phase very efficient.

5

Example 4:

The experiment of Example 3 was repeated, but now a vessel filled with glycerol was used instead. Similar results were obtained; bubbles of the same size rose from the 64
10 exits when air was blown through the horizontal structure. Again the bubbles rose in nice parallel streets without visible coalescence either horizontally or vertically.

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What is claimed is:

1. Method for operating a chemical and/or physical process in a vessel containing a liquid, a gas and/or solid particles, in which vessel a fluid is introduced through a hierarchical network of channels comprising parent and child generations of channel formations, wherein substantially each channel in a parent generation is divided into N channels of the child generation, whereby each of said child generations may in turn be a parent generation for a successive child generation, which network terminates in channel exits, such that said fluid is discharged from the channel exits substantially uniformly throughout the vessel volume.
2. Method according to claim 1 in which said network is a self-affine network of channels, wherein each of the channels in the parent generation have a diameter d_j and length l_j and each of the channels in the child generation have a diameter d_{j+1} and length l_{j+1} , wherein the ratios d_j/d_{j+1} and/or the ratios l_j/l_{j+1} are substantially constant for channels of successive generations running in parallel direction or in which method said network is a self-similar network which is a type of said self-affine network wherein the ratios d_j/d_{j+1} and l_j/l_{j+1} are substantially constant for channels of successive generations independent of their direction.
3. Method according to claim 2 wherein the ratios of diameters and/or lengths of channels in successive generations of said network are related to N by

$$N = (d_j/d_{j+1})^\Delta, \text{ and/or}$$

$$N = (l_j/l_{j+1})^D,$$
 wherein Δ and D represent an integer or a real positive number.

4. Method according to claim 3 wherein D is between 2 and 3.

5. Method according to claim 1, in which said chemical and/or physical process is selected from a group consisting of a fluidized bed process, a slurry process, an absorption process, a gas/liquid bubble column process and an aeration process.

6. Method for scaling up chemical and/or physical processes which processes are carried out in a vessel comprising the steps of building a small scale vessel in which vessel a fluid is introduced through a hierarchical network of channels comprising parent and child generations of channel formations, wherein substantially each channel in a parent generation is divided into about N channels of the child generation, which network terminates in channel exits, such that said fluid is discharged from the channel exits substantially uniformly throughout the vessel volume and determining optimal geometry and optimal values for the parameters N, Δ and D, and subsequently building a large scale vessel in which said geometry and parameters are substantially the same as in the small scale vessel.

7. Vessel containing a hierarchical network of channels, said network comprising parent and child generations of channel formations, wherein substantially each channel in a parent generation is divided into N channels of the child generation, whereby each of said child generations may in turn be a parent generation for a successive child generation, which network terminates in channel exits, wherein said network is a self-affine network of channels, wherein each of the channels in the parent generation have a diameter d_j and length l_j and each of the channels in the child generation have a diameter d_{j+1} and length l_{j+1} , wherein the ratios d_j/d_{j+1} and/or the ratios l_j/l_{j+1} are substantially constant for channels of successive generations running in

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parallel direction or in which vessel said network is a self-similar network which is a type of said self-affine network wherein the ratios d_j/d_{j+1} and l_j/l_{j+1} are substantially constant for channels of successive generations independent
5 of their direction.

8. Vessel according to claim 7 wherein the ratios of diameters and/or lengths of channels in successive generations of said network are related to N by

$$N = (d_j/d_{j+1})^A, \text{ and/or}$$

$$10 \quad N = (l_j/l_{j+1})^D,$$

wherein A and D represent an integer or a real positive number.

9. Vessel according to claim 8 wherein D is between 2 and 3.

15 10. Hierarchical network of channels comprising parent and child generations of channel formations, wherein substantially each channel in a parent generation is divided into N channels of the child generation, whereby each of said child generations may in turn be a parent generation for a
20 successive child generation, which network terminates in channel exits, which said network is a self-affine network of channels, wherein each of the channels in the parent generation have a diameter d_j and length l_j and each of the channels in the child generation have a diameter d_{j+1} and
25 length l_{j+1} , wherein the ratios d_j/d_{j+1} and/or the ratios l_j/l_{j+1} are substantially constant for channels of successive generations running in parallel direction or which network is a self-similar network which is a type of said self-affine network wherein the ratios d_j/d_{j+1} and l_j/l_{j+1} are substantially
30 constant for channels of successive generations independent of their direction and wherein the ratios of diameters and/or

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lengths of channels in successive generations of said network are related to N by

$$N = (d_j/d_{j+1})^{\Delta}, \text{ and/or}$$

$$N = (l_j/l_{j+1})^D,$$

5 wherein Δ and D represent an integer or a real positive number.

11. Network according to claim 10 wherein D is between 2 and 3.

10 12. Network according to claim 10 wherein at least one material is present near the exits of said network and/or said material is present as a coating on at least part of the inner surface of said network, which material is capable of chemical and/or physical interactions with fluids.

15 13. Network according to claim 10 which is provided with means for obtaining a gradient in the dimensions of the respective exits.

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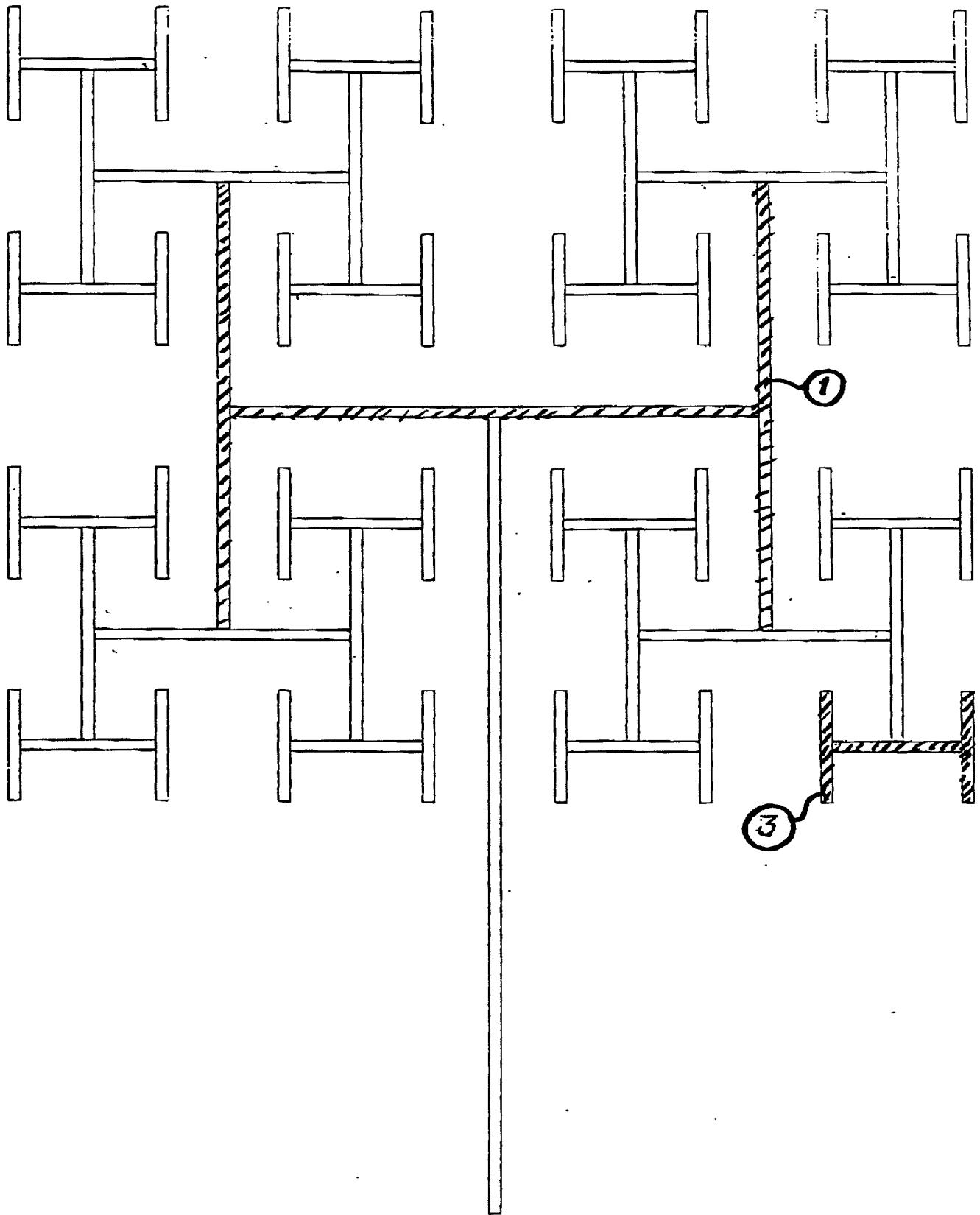


FIG.1

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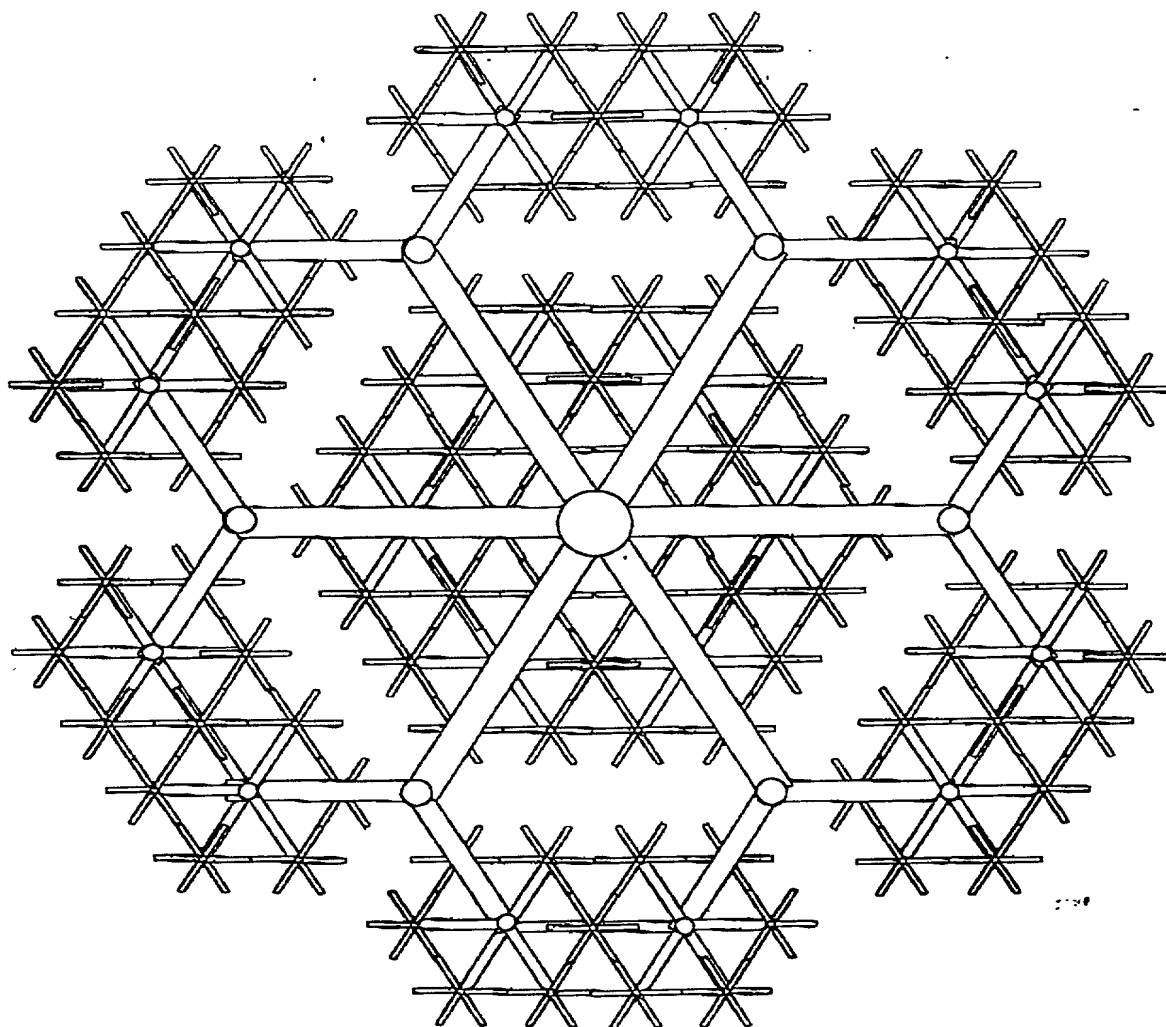
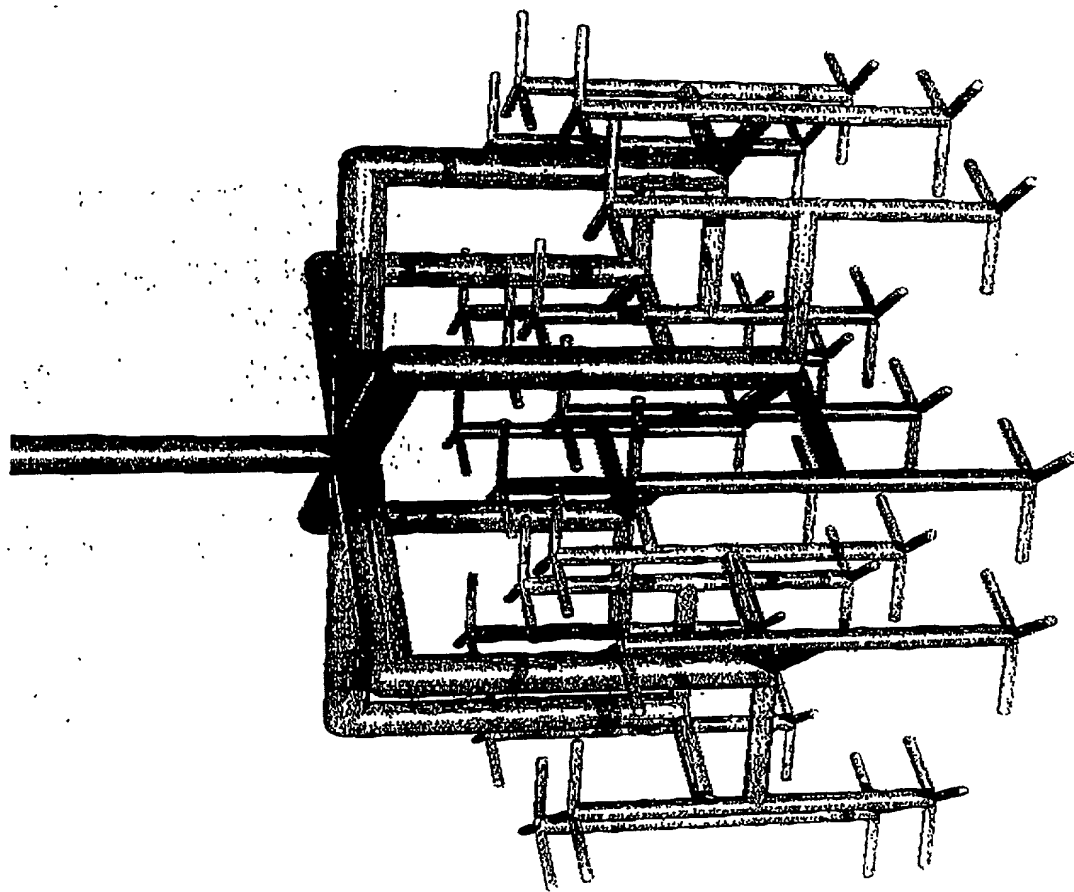


FIG. 2

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Figure 3



DECLARATION AND POWER OF ATTORNEY

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name;

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: METHOD FOR OPERATING A CHEMICAL AND/OR PHYSICAL PROCESS BY MEANS OF A HIERARCHICAL FLUID INJECTION SYSTEM

the specification of which (check one):

☒ is attached hereto. ☐ was filed _____ as Application No. _____
amended on _____ (if applicable).

☐ was filed as PCT International Application No. _____ on _____,
and was amended under PCT Article 19 on _____ (if applicable).

I ☒ hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I ☒ acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations §1.56(a).

I ☒ hereby claim foreign priority benefits under Title 35, USC §119(a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

<u>Prior Foreign Application(s)</u>	<u>Date Filed</u>	<u>Priority Claimed</u>	
_____ (Number) (Country)	_____ (Day/Month/Year)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number) (Country)	_____ (Day/Month/Year)	<input type="checkbox"/> Yes	<input type="checkbox"/> No

I hereby claim the benefit under Title 35, USC §119(e) of any United States provisional application(s) listed below:

_____ (Application Number)	_____ (Filing Date)
_____ (Application Number)	_____ (Filing Date)
_____ (Application Number)	_____ (Filing Date)
_____ (Application Number)	_____ (Filing Date)

Express Mail Number

EMS29469068US

I hereby claim the benefit under Title 35 USC §120 of any United States application(s) listed below and insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35 USC §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application No.)	(Filing Date)	(Patented/pending/abandoned)
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(Application No.)	(Filing Date)	(Patented/pending/abandoned)
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(Application No.)	(Filing Date)	(Patented/pending/abandoned)
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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) to prosecute this application and transact all business connected therewith in the Patent and Trademark Office, and to file with the USRO any International Application based thereon.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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